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Field/stress tunable behaviour of composites containing combined current-modulation annealed ferromagnetic microwires

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Polymer composites containing melt-extracted (MET) ferromagnetic microwires have demonstrated excellent sensing capabilities to magnetic bias or mechanical stress.[1,2] We have introduced an alternate/pulse current combined-modulation annealing (CCMA) to optimise MET wires' domain structure and enhance their giant magnetoimpedance (GMI) response significantly as compared to conventional annealing techniques.[3] Also, it has been confirmed that topological factors such as wire length play a crucial role in determining the electromagnetic (EM) properties of the wire-composites.[4] Herein, we present the field/stress tunable features of microwave behaviour of epoxy-based composites containing CCMA-ed Co-based microwires of different lengths so as to look into the effects of CCMA. A decrease-increase trend of permittivity (ε) spectra is revealed in the CCMA-ed wires composites in presence of magnetic fields. A linear positive stress-ε dependency is shown in the composite containing 25 mm CCMA-ed wires.

Experimentally, as-cast and CCMA-ed $Co_{69.25}Fe_{4.25}B_{12.5}Si_{13}Nb_1$ MET microwires with lengths of 15, 25 and 35 mm were embedded into Prime^{TM} 20 epoxy, followed by a standard curing cycle to realise wire-composite samples with dimensions 70×10×1.8 mm. The volume fraction of wires is fixed at 0.026 %. Ref. 3 details the technical parameters of CCMA. The *S*-parameters were measured in the frequency range of 0.3 to 6 GHz from a modified frequency domain spectroscopy with magnetic fields up to 1500 Oe and mechanical strain up to 2 %, respectively.[1,2] Using the Nicolson-Ross procedure for the transformation of the load impedance by a transmission line, the effective permittivity $\varepsilon = \varepsilon' - j\varepsilon''$ is determined by the transmission S_{21} and reflection S_{11} parameters.[5]An error analysis indicates the modest uncertainties in ε' (<5%) and ε'' (<1%) for the data.

From the zero-field permittivity dispersion (Fig. 1), it is observed an ε peak approximately at 4.0 GHz for the composite containing 15 mm CCMA-ed wires. It is well established that the dipole resonance arises at $f = c/2l\sqrt{\varepsilon_m}$,[6] where c=3×10^8 ms^{-1} and ε_m is the permittivity of the host matrix. By substituting l=15 mm and ε_m=6 for the epoxy matrix, we obtain a resonance frequency of 4.1 GHz, which corresponds to the observed resonance peak. The slight difference between these values can be attributed to the residual stress relaxation on the wires after the curing process, and the air gap in between the measuring setup and samples. Remarkably, this resonance peak is later gradually suppressed with external magnetic fields further increasing to 1500 Oe whereas another resonance peak emerges and blueshifts with magnetic bias increasing (Fig. 1). The identified peak blueshift is naturally related to ferromagnetic resonance of the microwires as per Kittel's equations.[7] The competition between these two effects yields such decrease-increase field tunable properties. Notably, this feature is unavailable in the composites containing as-cast wires (not shown here), indicating CCMA benefits EM response of wires due to the residual stress relaxation and a better-defined domain structure. Figure 2 displays the permittivity evolution with the applied external mechanical strain up to 2.0 %. In the composites containing 25 mm CCMA-ed wires, we identify an increase trend of the ε (Fig. 2). This dielectric contribution is because that the present annealing strategy has significantly improved the circumferential anisotropy of the wires, which significantly enhances the mechanical-dielectric coupling.[4] In another perspective, wire length is also at play in determining the dielectric behaviour of wire-composites. In the composite containing long microwires (35 mm), wires are preferentially entangled along longitudinal direction after curing process which in fact diminishes dielectric response because the electric component of incident waves is arranged along the sample width direction. On the other hand, in the composites containing short wires (15 mm), less intuitive trend is observed due to undesirable reflection loss arising from the more dipoles therein the composite. We conclude that CCMA would result in a large and controllable dielectric response to external stimuli, hence improving sensing performance of magnetic/mechanical sensors based on MET wires and their composites. All shown results suggest that the polymer composites enabled by CCMA-ed microwires are promising in magnetic/stress sensors.

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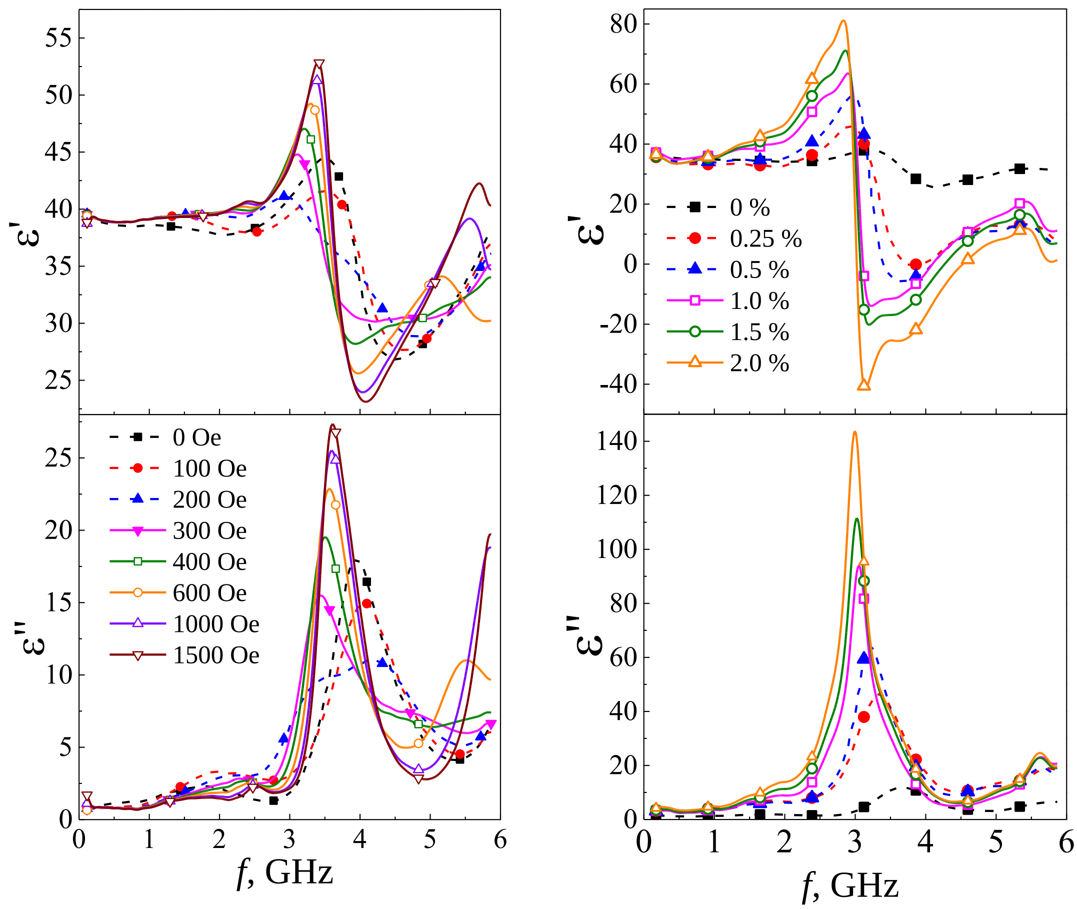

Fig. 1 Frequency dependency of permittivity of composite containing 25 mm CCMA-ed microwires with magnetic fields up to 1kOe

Fig. 2 Stress tunable behaviour of the composite containing 25 mm CCMA-ed microwires with mechanical strain up to 2.0 %